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Natural and Biological Inspiration to Remediate Sewage Water for Irrigation

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ABSTRACT
Natural cleanup of sewage water has great attention in the recent years. Natural remediation and bioremediation were tested to reduce the heavy metal pollutants of sewage water which collected from El-Suff district, Giza Governorate, Egypt (29 º 34' 38'' N latitude and 31º 17' 26'' E longitude). The treatments were Biochar, Zeolite, Cyanobacteria (*Trichormus variabilis)* and a mixture of all tested materials. The treated samples were subjected to determine total dissolved solids (TDS), N-NO_3, phosphorus, biological oxygen demand (BOD), chemical oxygen demand (COD) and some heavy metals (Fe, Cd and Ni) 10, 20 and 30 days to assess its suitability for irrigation use. All treatments demonstrated an effective clean-up activity and decreasing the concentration of heavy metals, with the mixture of three tested materials that being the most successful, followed by zeolite and biochar then, cyanobacteria *Trichorumus variabilisas* individual treatments.

Keywords: Swage water, Natural remediation, Bioremediation, Cyanobacteria, *Trichorumus variabilisas*, Zeolite, Biochar.

1. Introduction

Pollution is considered as one of the biggest challenges to humanity. So, research is being conducted to execute the notion of pollutants removal from wastes. Water is one of the most impacted environmental aspects, as it is constantly polluted with heavy metals from many sources (Visa, 2016).

Access to a clean and affordable water supply (SDG Goal 6) is one of the sustainable development goals (SDGs) adopted by the United Nations in 2015, with Target 6.1: By 2030, achieve universal and equitable access to safe and affordable drinking water for all. Increased urbanization has an impact on water quality because pollutants generated in cities settle and pollute water and because cities, industries, and agriculture are competing for more water (Zhang *et al.,* 2021)

There are several approaches to transform wastewater environmental issues into effective, commercially feasible techniques of energy production and water recycling. Recycling wastewater is so important for water conservation (Abedi *et al.,* 2019). Generally, physical and chemical remediation procedures for the removal of organic contaminants are commonly used. Although these approaches are considered appropriate for removing organic contaminants from the environmental sites, but they are expensive and difficult to apply (Selvi *et al.,* 2019). Bioremediation is an environmentally beneficial method of cleaning up contaminated soil and polluted water. Biological treatment can be achieved in a variety of methods, including the use of mixed microbial cultures such as bacteria, fungus and algae for pollutant bioremediation (Azab, 2008).

Biochar made from agricultural residual is becoming more widely recognized as a versatile material for several purposes (Yakout, 2015). It is a complex carbonaceous matrix that can be produced easily and cheaply by pyrolyzing bio wastes under oxygen-limited conditions (Shaheen *et al.,* 2019). Biochar produced from bio waste is gaining popularity as an economical and sustainable

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resource with numerous environmental applications (Bandara *et al.,* 2020). It has been extensively researched and has prospective applications in agriculture, environment, and energy to provide food security (Li *et al.,* 2019) clean environment and adequate energy due to its low cost, wide availability and eco-friendliness (He, *et al.,* 2018). Biochar is being used as a bio-sorbent for the removal of toxic metal (Li *et al.,* 2020), and other emerging contaminants such as antibiotics. Thus the removal efficiency of emerging pollutants can be improved through some modification (e.g., combined biochar with nanotechnology or microorganism) to improve the electron donor ability of biochar (Yuan *et al.,* 2017).

Zeolites play an important role in the conversion of solid and liquid hazardous wastes into environmentally acceptable products (Shevade and Ford, 2004). Natural zeolites are hydrated alumino-silicate materials with remarkable ion-exchange and sorption capabilities that are both environmentally and economically acceptable. Their efficiency in various technological processes is determined by their physical-chemical qualities, which are inextricably linked to their geological deposits. Natural zeolites have a wide range of applications due to their distinctive tree dimensional porous structure. Natural zeolites are cationic exchangers due to an excess of negative charge on the surface of the zeolite caused by isomorphic replacement of silicon by aluminum in the basic structural units. Numerous investigations have demonstrated their remarkable efficacy in removing metal cations from wastewaters (Margeta *et al.,* 2014).

Phyco-remediation has been utilized to treat municipal wastewater as a cost-effective nutrient-removal technology utilizing microalgae since around 1963, from the advantages of phyco-remediation, removal, transformation, or degradation of nutrients, organic matter, acids, metals and xenobiotic compounds, as well as the use of algae as an environmental monitoring system for toxic material detection and the production of high-value metabolites (Esquivel-Hernández. *et al.,* 2017).

Cyanobacteria (*Trichorumus variabilisas*) are unusual creatures with a high degree of environmental adaption. They may respond to various forms of organic pollutants in various ways, ranging from bioaccumulation to biodegradation; they have an enormous potential to adapt and grow in environments with low fluid potential. In arid environments, they can tolerate salinity and they can grow in hypersaline environments. Cyanobacteria *Trichorumus variabilisas* can successfully be employed to remediate contaminated environments due to their diverse metabolism and ability to quickly convert from one phase of growth to another (Zinicovscaia and Cepoi, 2016). *Trichormus variabilis* in terms of the various types of bioenergy and nutrient removal potential, synonyms of *Trichormus variabilis*, such as Anabaena variabilis (Abedi *et al.,* 2019).

The study aims to assess the effectiveness of using zeolite, biochar and cyanobacteria (*Trichormus variabilis*) as remediation agents to remediate sewage water even separately or in combination.

2. Materials and Methods

The study was carried out in Soils, Water and Environment Research Institute, Agricultural Research Center, Giza, Egypt. Samples of sewage water were collected from El-Suff district, Giza Governorate, Egypt (29 º 34ˉ 38⁼ N latitude and 31 º 17ˉ 26⁼ E longitude).

2.1. Natural and Biological remediation agents

Zeolite**,** Biochar and *Trichormus variabilis* cyanobacterial strain along with were supplied by Agric. Microbiology and Chemical & Physical Departments, Soils, Water and Environ. Res. Inst., ARC, Giza, Egypt.

2.2. Experimental Design

Chemical and biological analysis for sewage water

The water assay were (pH), total dissolved solids (TDS), nitrate (NO_3), phosphorous (P), biological oxygen demand (BOD), chemical oxygen demand (COD), Cadmium (Cd), Ferric (Fe) and Nickel (Ni) were measured before starting the experiment (initial analysis) and every 10 days for three times, also assess. The difference between the initial and final values provided an estimate of microalgae phyco-remediation.

100 ml of sewage water was put it in suitable conical flasks. Each flask received one of the following materials: 10 g zeolite, 10 g biochar, 10 ml cyanobacteria ($6 * 10^2$ CFU/ml) and a mixture of all previous materials, while the control flask received none of tested materials (five treatments). Each treatment was replicated three times (total of 15 flasks). Every ten days, one flask from each treatment was taken and analyzed.

Microalgae *Trichormus variabilis* cells were subculture in BG_{11} media prior to reaching the stationary phase. Before using it for microalgae culture, BG_{11} medium was sterilized in an autoclave for 15 minutes at 121ºC.

3. Results and Discussion

3.1. Initial chemical properties of sewage effluent

Table 1 shows some initial chemical properties of sewage effluent before treatments. Schwarzenbach *et al.* (2010) reported that the chemical parameters including pH, TDS, TSS, BOD, COD, nutrients and heavy metals indicate the wastewater quality. The clean and wastewater had BOD values of 2 and 10 mg L^{-1}, respectively, The BOD value of sewage effluent under study was exceeded than limits of the Egyptian Law (48/1982) for wastewater (Isukul *et al.,* 2023). The primary factors for determining irrigation water quality are pH and TDS. The chemical parameters are important indicator to evaluate wastewater pollution of organic and inorganic materials contents (Eman *et al.,* 2023) and reuse it with quality criteria for crops irrigation (FAO, 2015).

Table1: Some chemical properties of sewage effluent

pH	SAR	COD	BOD	TSS	TDS	PO_4	NO_3	Ca	Mg	Na	Cu	Cd	Fe	Ni
		mgO_2L^{-1}		mgL^{-1}								μgL^{-1}		
7.4	12.4	94.3	65.6	18.4	379.8	2.43	0.97	39.6	15.5	79.6	32.6	25.8	318.5	51.4

SAR: sodium adsorption ratio, COD: Chemical oxygen demand, BOD: biological oxygen demand, TSS: total suspended solids, TDS: total dissolved solids

3.2. Effect of different treatments with natural materials on sewage effluent after 10 days

Bio and natural treatments were found to be a positive effective on sewage effluent properties (pH, TDS, NO3, P, BOD, COD, Cd, Fe and Ni)

The results in Table 2 showed that all treatments under study led to decrease in wastewater chemical parameters values after 10 days, but the more evident with combination of all treatments. Almost, the trend of treatments as following: the combination of all treats > zeolite treat > biochar treat> *Trichormus variabilis* treat, while zeolite treat was the better to reduce Na, Cu, Cd, Fe and Ni.

Table 2: Some chemical properties of sewage effluent after 10 days treatment

Treatments	pH	SAR	COD	BOD	TSS	TDS	PO_4	NO_3
			mgO_2L^{-1}		mgL^{-1}			
Zeolite	7.2a	10.79c	59.6c	57.5b	11.7d	271.6c	1.81c	0.50d
Biochar	7.3a	11.68b	63.2c	59.3a	12.5c	280.1b	1.95a	0.59a
Trichormus variabilis	7.3a	12.22a	72.5b	60.2a	13.6b	286.5a	1.77d	0.54c
Mix of all treats	7.2a	10.31c	57.3d	55.8c	9.8e	267.3d	1.61e	0.49d

Table 2: Continued

Treatments	Ca	Mg	Na	Cu	Cd	Fe	Ni
	mgL^{-1}				μgL^{-1}		
Zeolite	30.5c	11.7c	51.8d	9.5e	10.3d	196.8c	28.8d
Biochar	33.8b	12.2b	55.5c	11.4d	12.8c	207.5b	31.3c
Trichormus variabilis	33.6b	12.8a	56.3b	12.3c	14.5a	210.3b	33.3b
Mix of all treats	32.4b	11.2c	52.6c	10.2d	10.6d	198.6c	30.5c

SAR: sodium adsorption ratio, COD: Chemical oxygen demand, BOD: biological oxygen demand, TSS: total suspended solids,
TDS: total dissolved solids
Means within each line followed by the same letter are not significantly different ($p<0.05$)

Also, it was clear from the previous Table that the *Trichormus variabilis* treatment was better to reduce PO4 concentration in sewage effluent than zeolite and biochar treatments with values were 1.77, 1.81 and 1.95mgL^{-1}, respectively, but the mix of all treats was the best with value 1.61mgL^{-1}. The results revealed that all treatments decrease both nitrogen and phosphorous concentration in sewage effluent and it's agreed with those obtained by Sharma *et al.* (2020). *Trichormus variabilis* have long been employed as low-cost remediates for phosphorus (P) and nitrogen (N)-rich residential wastewaters, converting these nutrients into biomass (Singh *et al.,* 2011). Microalgae can utilize N and P present in the wastewater as nutrients source to support its growth. Nitrogen is absorbed by algae in the form of nitrate and ammonia, whereas phosphorus is absorbed in the form of phosphoric acid (Lam *et al.,* 2017).

3.2. Effect of different treatments with natural materials on sewage effluent after 20 days

Chemical oxygen demand (COD) and biological oxygen demand (BOD) are good indicators of the amount of organic contaminants in wastewater. Wastewater with high BOD and COD levels limits the availability of soluble oxygen to aquatic life. So, before releasing wastewater into fresh water bodies, BOD and COD loads should be lowered.

Table 3: Some chemical properties of sewage effluent after 20 days treatment

Treatments	pH	SAR	COD	BOD	TSS	TDS	PO_4	NO_3
			mgO_2L^{-1}		mgL^{-1}			
Zeolite	7.1a	8.66c	53.5c	48.8c	8.6c	182.2c	1.51b	0.41b
Biochar	7.2a	10.51b	57.4b	51.7b	11.3b	204.5b	1.66a	0.52a
Trichormus variabilis	7.4a	13.56a	62.6a	55.8a	13.5a	221.4a	1.68a	0.55a
Mix of all treats.	7.2a	9.22c	52.8c	46.5c	8.5c	178.6d	1.31c	0.39b

Table 3: Continued

Treatments	Ca	Mg	Na	Cu	Cd	Fe	Ni
	mgL^{-1}				μgL^{-1}		
Zeolite	26.8b	10.2b	46.5b	7.3d	7.6c	150.3b	19.6c
Biochar	29.3a	11.5a	50.2a	9.8c	9.7b	198.2a	23.5b
Trichormus variabilis	28.5a	11.6a	51.4a	11.5b	11.2a	196.5a	29.5a
Mix of all treats.	23.6c	9.8b	45.3b	8.5c	8.3b	152.8b	17.7c

SAR: sodium adsorption ratio, COD: Chemical oxygen demand, BOD: biological oxygen demand,
TSS: total suspended solids, TDS: total dissolved solids
Means within each line followed by the same letter are not significantly different (p<0.05)

The results in Table 3 showed that after 20 days of treatment, the same previous trend was in reducing pollutant concentrations. The results showed that all tested treatments were clearly effective in reducing the concentration of nutrients, microelements, and heavy metals from sewage effluent under study (El-Hameed Mona *et al.,* 2021). Heavy metals that polluted water are significant environmental problem (Ramadan *et al.,* 2020). Heavy metals in wastewater can be handled with a range of technologies, including chemical, physical and biological methods (El-Bestawy, 2019).

The biochar treatment recording a positive effect to removal of all heavy metals because of its surface heterogeneity, biochar might have a high sorption capacity for metallic pollutants, similar to activated carbon (Kasozi *et al.,* 2010). Due to metallic ions can be physically sorbed onto the surface of the char and held within the pores, biochar with high surface areas and pore volumes have a high affinity for metals absorbance (Kumar *et al.,* 2011). Various metals can interact with specific ligands and functional groups on biochar to form complexes (Wang *et al.,* 2015) or precipitates of their solid mineral phases (Inyang *et al.,* 2012). Algae and cyanobacteria prefer to grow on heavy metals present in wastewater, so absorb it with large amount (Wan Maznah *et al.,* 2014).

3.3. Effect of different treatments with natural materials on sewage effluent after 30 days

After 30 days, all treatments showed a considerable decrease in all chemical parameters of sewage effluent under study especially with zeolite and biochar treatments.

The results in Table 4 showed a decrease in the COD and BOD values after 30 days of treatment, with values of 50.4 and 45.6, 52.7 and 50.2, 66.8 and 53.6 and 54.3 and 48.3 mg $O_2L^{-1}$for zeolite, biochar, *Trichormus variabilis* and mix treatments, respectively. The effect of biochar removal efficiency on COD, BOD, and TDS was became increase more obvious with time. This finding was consistent with that obtained by Karthikeyan *et al.* (2019). Currently, biochar is a promising material as an adsorption agent of toxins, which could also help reduce water pollution (Tan *et al.,* 2015).

Salgueiro *et al.* (2016) found that an increase in the growing period would increase the concentration of microalgae cells, which can be ascribed to increased glucose uptake and COD and BOD elimination. In general, organic matter sorption on biochar is a complex process that is regulated by polarity, hydrophobicity, aromaticity, and molecular size (Nguyen *et al.,* 2007).

Table 4: Some chemical properties of sewage effluent after 30 days treatment

Treatments	pH	SAR	COD	BOD	TSS	TDS	PO_4	NO_3
			mgO_2L^{-1}		mgL^{-1}			
Zeolite	7.1a	7.48d	50.4b	45.4c	7.4c	171.5d	1.46d	0.37d
Biochar	7.1a	9.89c	52.7b	50.2b	9.5b	193.2b	1.60b	0.48b
Trichormus variabilis	7.5a	15.33a	66.8a	53.6a	14.6a	226.8a	1.71a	0.58a
Mix of all treat.	7.3a	10.41c	54.3b	48.3b	9.2b	185.4c	1.53c	0.40c

Table 4: Continued

Treatments	Ca	Mg	Na	Cu	Cd	Fe	Ni
	mgL^{-1}				μgL^{-1}		
Zeolite	23.3b	10.0b	41.3c	6.4c	5.9c	132.5e	15.5c
Biochar	27.5b	10.1b	47.6b	7.5c	8.5b	163.8c	18.9b
Trichormus variabilis	31.2a	12.3a	53.5a	10.8b	10.6a	185.4b	24.3a
Mix of all treat.	26.8a	11.4b	46.5b	7.9c	7.9b	150.2d	16.9b

SAR: sodium adsorption ratio, COD: Chemical oxygen demand, BOD: biological oxygen demand,
TSS: total suspended solids, TDS: total dissolved solids
Means within each line followed by the same letter are not significantly different (p <0.05)

Furthermore, use zeolite, biochar, mix of all treatments and *Trichormus variabilis* led to decrease of COD values with percentage as follows: 46.6, 44.1, 42.4 and 29.2%, respectively, and were reduced BOD values with percentage as follows 30.8, 23.5, 26.4 and18.3%, respectively, compared to the initial data. Zeolite was the best treatment, which led to decrease COD and BOD values after 30 days. Because of the steric hindrance of different molecule sizes, the effective contact and interception effects on biological carbon vary and thus the adsorption process and adsorption impact differ. The treatment of contaminated water with microbial/photosynthetic bacteria and microalgae-bacteria media significantly degrades organic matter and eliminates COD and BOD with effective effect (Sheng *et al.,* 2012). The development of low-cost and effective ecological engineering approaches, as well as the selection or natural growth of specific bacteria is important to the success of this process. The high rate of COD and BOD removal indicating that microalgae species can survive and thrive at high COD and BOD levels (Wang *et al.,* 2012).

Use of natural materials in treating polluted water with adsorption mechanism is considered an effective method (Tasić *et al.,* 2019). Using zeolite achieved decreasing of COD and BOD (Zheng *et al.,* 2015). The organic molecules from the wastewater trapped in the pores of zeolite could be responsible for the reduction of COD and BOD (Fanta *et al.,* 2019). Zeolite features (ion exchange capability, large surface area) have also been used in many biological processes. Zeolite have a three-dimensional structure made up of (Si, Al) O_4 tetrahedral connected at all of their oxygen vertices. These channels allow for the exchange of cations and H_2O molecules to balance the negative charge left over from the isomorphous substitution. Because the aluminum ion is so tiny, take up the position in the four-sided tetrahedron's center, the substitution of Al^{3+} for Si^{4+} and the presence of oxygen atoms produce a Lattice's negative charge is balanced by the interchangeable replacement of the cation (sodium, potassium, or calcium) of heavy metals (Yuna, 2016). The exchange of Fe, Cd and Ni ions with zeolite was described by Hong *et al.,* (2019) whom found that while the process for copper, lead and nickel exchange was related to the cation exchange capabilities of the zeolite framework, the mechanism for metal ion uptake was related to pore design.

Irrigation water with high TDS causes salt deposition and increases soil water osmotic potential, reducing crop water availability and productivity (Zaman *et al.,* 2018). Total dissolved solids (TDS) are the total quantity of metals, cations, anions, minerals and salt those have been dissolved in water. To avoid secondary soil salinization, the TDS value of irrigation water should be less than 250mgL^{-1}. The initial TDS value of sewage effluent was 379.8 mgL^{-1}, and after 30 days of tested natural materials, TDS values were reduced significantly. The zeolite treatment showed evidence decrease (171.5 mg/L), followed by mixture of all treatments (185.4 mg/L). The total dissolved solids (TDS) were reduced by 54.9 and 51.2% for zeolite and mix of all treatments, respectively, compared to the initial data of sewage effluent. The zeolite material is more effective for reducing the TDS in wastewater. *Trichormus variabilis* cyanobacterial strain was effective to reduce TDS in sewage effluent according to Khan *et al.,* (2019). They stated that TDS was reduced due to the consumption of dissolved solid from wastewater, which is nutrition rich for the growth of microalgae (Malla *et al.,* 2015). Because of the presence of diverse agents, the combination of treatments demonstrated realized the utmost reduction in COD, BOD and TDS. These results agreed with those obtained by Wu *et al.* (2009) whom reported that the employment of microbial technology in conjunction with other techniques has shown reasonable efficacy in the remediation of polluted water.

4. Conclusion

Biochar, zeolite and cyanobacteria (*Trichormus variabilis*) might be effective remediation agents of sewage water. Zeolite and biochar are physical agents, whereas Trichormus variabilis is a biological agent and all of them can be used to purify sewage water as individual treatments. However, mixture of these materials are advised in sewage water remediation to increase the likelihood that the wastewater can be used in the irrigation process. More research is needed to optimize the use of diverse concentrations of zeolite and biochar, as well as duration in wastewater treatment experiments.

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